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ASSESSMENT OF MICRO-AGGREGATION USING LASER DIFFRACTOMETRY

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ABSTRACT

In order to evaluate the influence of the measuring technique on the determination of (micro-)aggregation in soil and sediment samples, results of grain size distributions of undispersed silty soil samples obtained by the sieve–pipette method are compared with those obtained using a laser diffraction grain size analyser, the Coulter LS-100. Reduced major axis relationships are calculated which may be used to convert Coulter LS-100 results to those obtained by the sieve-pipette method. The relationships obtained are very similar to the reduced major axis relationships established for dispersed silty soil samples. The results also show that the Coulter LS measurements have a systematic bias compared to the sieve–pipette data. This implies that, if the percentage of (micro-)aggregation is determined, the (interpretation of the) results will be strongly dependent on the measurement technique used. Using the calibration relationships that were established, nomographs can be developed to predict the level of sieve–pipette (micro-) aggregation from Coulter LS-100 data. Copyright © 1999 John Wiley & Sons, Ltd

KEY WORDS: micro-aggregation; aggregate size distribution; laser diffraction; sieve–pipette method

INTRODUCTION

Slattery and Burt (1995) made a distinction between the ultimate and the effective size distribution of a sediment sample. The ultimate (i.e. dispersed) size distribution is most important with respect to certain soil chemical and soil physical properties. However, it is clear that the effective (i.e. undispersed) size distribution of the sediment plays a major role in processes like sediment transport by wind or water (e.g. Meyer *et al.*, 1992; Walling, 1990). Slattery and Burt (1995) stated that most of the sediment eroded from agricultural soils is composed of aggregated particles.

Although the sieve–pipette method is still the standard method for grain size analysis of dispersed soil samples, laser diffraction is increasingly used worldwide. Various authors (e.g. Loizeau *et al.*, 1994, Konert and Vandenberghe, 1997) have compared grain size analyses by the sieve–pipette method with the laser diffraction method for dispersed soil samples. In a recent study Beuselinck *et al.* (1998) proposed reduced major axis–relationships to convert grain size fraction obtained by the Coulter LS-100 to those obtained by the sieve–pipette method for silty soil samples.

Various methods have been used to determine the size distribution of aggregated sediments. Nicholas and Walling (1996) used a field-based elutriation system based on the same principle as the pipette method to investigate the effects of suspended sediment aggregation on rates of overbank deposition and on the grain size composition of deposited sediment. Lick (1982) measured the effective size distribution

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using a settling tube. Slattery and Burt (1995) used a combination of the sieve–pipette method and laser granulometry to determine the dispersed and the undispersed particle sizes of eroded sediment. Lau and Krishnappan (1994) determined the undispersed particle sizes of cohesive sediment by laser granulometry. Muggler *et al.* (1997) stressed the potential of a laser diffractometer to measure aggregation in soil science.

There is no standard method for measuring the effective size distribution of soils and sediments. Laser diffractometry shows great promise as it is fast, requires only minute amounts of sediment and has a wide measuring range (0·4-4000 μm). The aim of this study is, first, to determine if the effective particle size distribution measured by a laser diffractometer is comparable to the effective particle size distribution measured by the sieve–pipette method, and second, to determine the implications of using one of the two methods for the assessment of sample aggregation.

MATERIALS AND METHODS

Principles of the Coulter LS-100

Laser diffractometry size analysis is based on the principle that particles of a given size diffract light through a given angle, the angle increasing with decreasing particle size. A narrow beam of monochromatic light ($\lambda = 750$ nm) is passed through a sample cell containing an upward-moving suspension. The diffracted light is focused onto 126 detectors. To calculate the grain size distribution from the light intensity reaching the array of detectors, the Fraunhofer theory is used (de Boer *et al.*, 1987). The Coulter LS-100 measures the volumetric percentage in 72 classes ranging from 0·4 to 900 μm, corresponding to an interval of 0·155ϕ ($\phi = -\log_2 D$, where D = the grain size in mm). Degassed, distilled water is used as suspension fluid. Loizeau *et al.* (1994) discuss the mechanisms involved in more detail.

Principles of the sieve–pipette method

This method is a combination of wet sieving of the fraction > 63 μm and the pipette sampling method, based on Stokes' law, for the silt (2–63 μm) and clay (< 2 μm). In a column the sediment concentration as a function of time is monitored by timed withdrawals of pipette samples at a certain height and at a constant temperature. Sediment concentrations are measured at combinations of time and depth corresponding to particle diameters of 32, 16, 8, 4 and 2 μm (i.e. at 1ϕ intervals). The sieve–pipette method measures the mass percentage for the defined grain size classes.

Samples and sample preparation

A total of 81 soil samples were analysed by both methods before and after dispersion. Soil samples were taken at various locations in a small study area located 10–15 km to the southwest of Leuven, Belgium. Dominant soils in this area are loess-derived luvisols. The grain size distribution of the natural soil samples showed relatively little variation. M·ore extreme variations in grain size distribution were therefore obtained using two procedures: (i) clay-rich soil samples were prepared by suspending a natural silty soil sample in distilled water by gentle stirring and removing the material still in suspension after a certain time interval; (ii) sand-rich silty soil samples were prepared by mixing a natural silty soil sample in known proportions with sand obtained by sieving another subsample of the same soil. All particle size analyses were conducted without removal of organic matter from the samples, as this would have destroyed aggregates present in the sediment (Slattery and Burt, 1997).

Table I. Reduced major axis (RMA) relationships between the Coulter LS-100 method and the sieve–pipette method for the different grain size classes and the 20, 50 and 80 percentiles of undispersed silty soil samples

Size class (μm)	Number of samples	RMA*	Correlation coefficient	Standard error of RMA slope	Critical percentage†	Threshold percentage‡
> 63	81	$y = 1{\cdot}120x – 6{\cdot}350$	0·98	0·025	5·67	52·92
2–63	81	$y = 1{\cdot}247x – 20{\cdot}034$	0·73	0·095	16·07	81·11
32–63	81	$y = 1{\cdot}222x – 3{\cdot}653$	0·94	0·047	2·99	16·45
16–32	81	$y = 1{\cdot}552x – 7{\cdot}058$	0·93	0·063	4·55	12·79
8–16	81	$y = 0{\cdot}988x – 1{\cdot}655$	0·93	0·040	1·68	–
4–8	81	$y = 1{\cdot}018x – 2{\cdot}107$	0·92	0·045	2·07	–
2–4	81	$y = 0{\cdot}858x – 1{\cdot}105$	0·96	0·025	1·29	–
< 2	81	$y = 2{\cdot}646x – 7{\cdot}782$	0·97	0·074	2·94	4·73
All classes	168	$y = 1{\cdot}227x – 3{\cdot}251$	0·90	0·022	2·65	14·32
20% >	67	$y = 0{\cdot}994x – 4{\cdot}955$	0·99	0·022	4·99	–
50% >	67	$y = 0{\cdot}954x – 1{\cdot}479$	0·98	0·022	1·57	–
80% >	67	$y = 1{\cdot}227x – 2{\cdot}504$	0·86	0·080	2·04	11·04

* x = Coulter LS-100 volume (%), y = sieve–pipette mass (%); for 20% >, 50% > and 80% >, x = Coulter LS-100 grain size (μm), y = sieve–pipette grain size (μm)
† Critical percentage (grain size (μm)) = x at which y equals 0
‡ Threshold percentage (grain size (μm)) = x at which y equals x

RESULTS

Since both measurements are subject to errors, the reduced major axis (RMA line) was calculated to relate sieve–pipette mass percentages and the Coulter LS-100 volumetric percentages measured for the different size classes. The RMA procedure minimizes the product of the deviations in the X and the Y directions, attributing the scatter of the data points to both variables. The slope of the RMA line is defined as the ratio of the standard deviations of the two variables. For each RMA line the correlation coefficient and the standard error of the RMA slope were calculated (Davis, 1986).

All RMA equations for the undispersed silty soil samples are significant at a 1 per cent level with correlation coefficients varying between 0·73 and 0·98 (Table I). Correlation is good for the clay, the sand and silt fractions. A weaker relationship is found for total silt fraction (2–63 μm). The slope is especially steep for the clay fraction. For most of the fractions the Coulter LS-100 method overestimates the grain size percentage up to a certain threshold, then underestimates it at higher percentages (Table I). For the clay fraction (< 2 μm) the relative overestimation by the Coulter LS-100 method is limited to small percentages. On the other hand, the sand fraction and coarse silt fractions are overestimated up to higher percentages. For the finer silt fractions (2–4 μm, 4–8 μm and 8–16 μm) the Coulter LS-100 always overestimates the size percentage. Beuselinck *et al.* (1998) discuss the possible reasons for the under- and overestimation of grain size percentages by the Coulter LS-100.

The overestimation implies that below a certain grain size percentage measured by the Coulter LS-100, negative sieve–pipette values would be calculated using the RMA relationships. It is suggested that the Coulter LS-100 percentage of that grain size class can be used as the sieve–pipette percentage for percentages smaller than this critical value (Table I), since small percentages are measured by both methods.

The slopes of the RMA lines of the 20 and 50 percentiles are close to 1, with high correlation coefficients, while the 80 percentile has a slightly steeper slope. For the 80 percentile the data are also more scattered than for the 20 and 50 percentiles.

Table II. Reduced major axis (RMA) relationships between the Coulter LS-100 method and the sieve–pipette method for the different grain size classes and the 20, 50 and 80 percentiles of dispersed silty soil samples (Beuselinck *et al.*, 1998)

Size class (μm)	Number of samples	RMA*	Correlation coefficient	Standard error of RMA slope	Critical percentage†	Threshold percentage‡
>63	83	$y=1{\cdot}155x–6{\cdot}105$	0·98	0·025	5·29	39·39
2–63	83	$y=1{\cdot}384x–35{\cdot}987$	0·73	0·104	26·00	93·72
32–63	83	$y=1{\cdot}230x–0{\cdot}671$	0·95	0·042	0·55	2·92
16–32	83	$y=1{\cdot}514x–7{\cdot}535$	0·92	0·065	4·98	14·66
8–16	83	$y=0{\cdot}869x–2{\cdot}746$	0·74	0·064	3·16	–
4–8	83	$y=0{\cdot}660x–1{\cdot}411$	0·93	0·027	2·14	–
2–4	83	$y=0{\cdot}805x–1{\cdot}611$	0·94	0·030	2·00	–
<2	83	$y=2{\cdot}744x–7{\cdot}773$	0·98	0·060	2·83	4·46
All classes	581	$y=1{\cdot}394x–5{\cdot}275$	0·86	0·030	3·78	13·39
20% >	68	$y=1{\cdot}025x–6{\cdot}474$	0·99	0·054	6·32	–
50% >	68	$y=1{\cdot}077x–3{\cdot}488$	0·99	0·018	3·24	45·30
80% >	68	$y=1{\cdot}563x–2{\cdot}306$	0·90	0·027	1·48	4·10

* x = Coulter LS-100 volume (%), y = sieve–pipette mass (%); for 20% >, 50% > and 80% >, x = Coulter LS-100 grain size (μm), y = sieve–pipette grain size (μm)
† Critical percentage (grain size (μm)) = x at which y equals 0
‡ Threshold percentage (grain size (μm)) = x at which y equals x

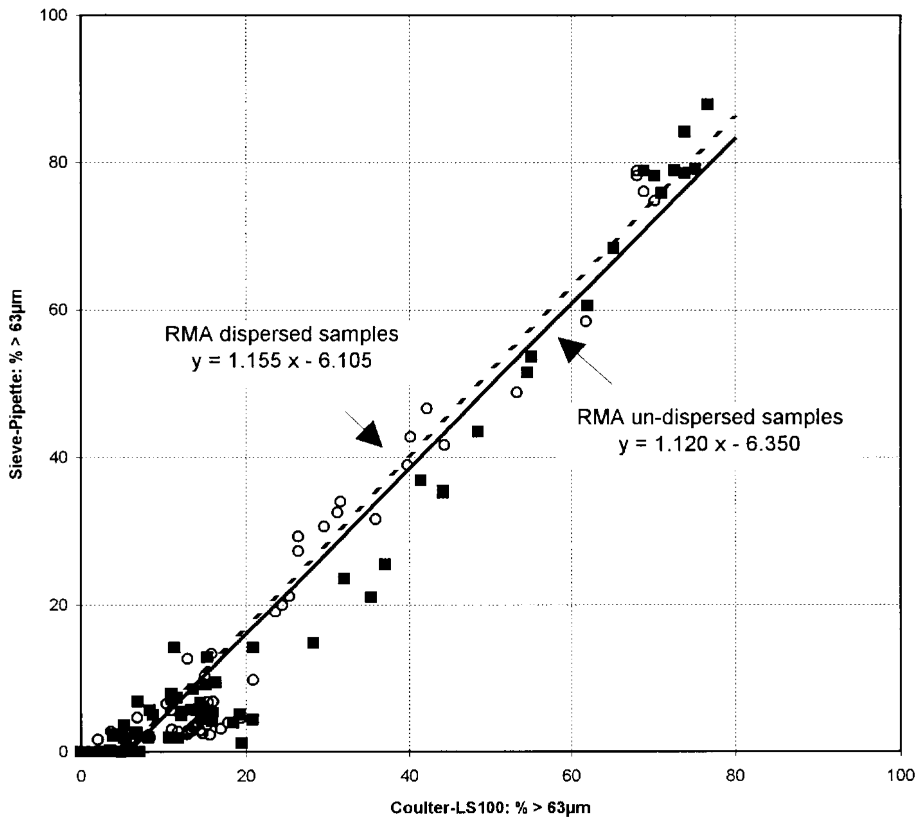


Figure 1. Comparison of the reduced major axis (RMA) relationships between the Coulter LS-100 and the sieve–pipette method for the sand fraction (>63 μm) of dispersed and undispersed silty soil samples (see Tables I and II for statistical details). o, Dispersed silty soil samples (Beuselinck *et al.*, 1998); ■, Undispersed silty soil samples; _____, RMA relationship for undispersed samples; - - - - -, RMA relationship for dispersed samples

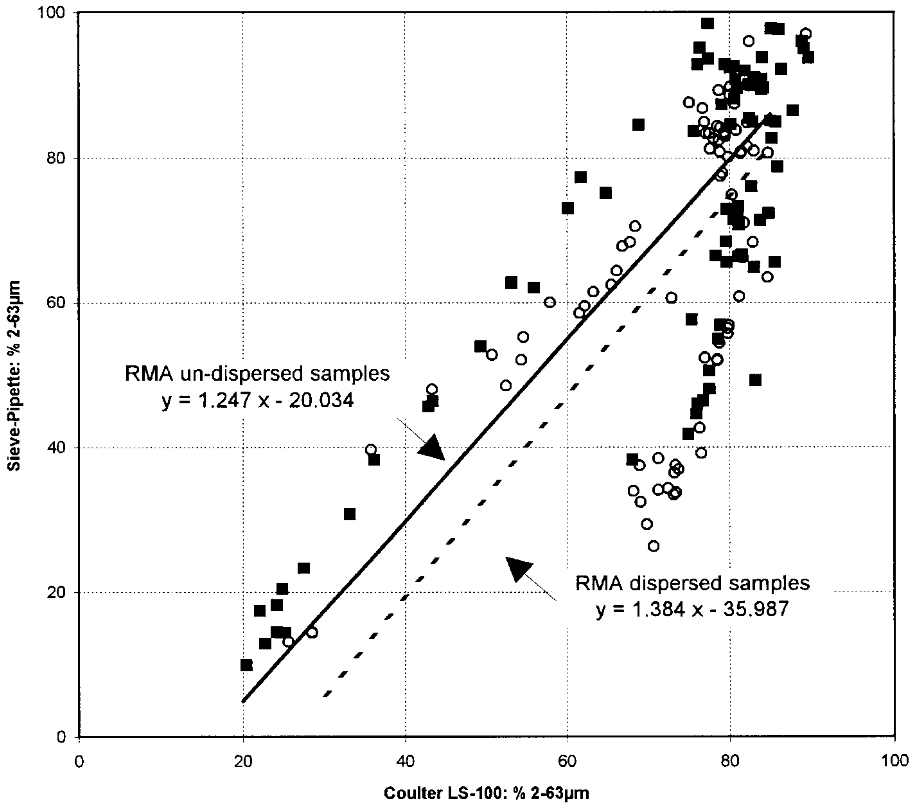


Figure 2. Comparison of the reduced major axis relationships between the Coulter LS-100 and the sieve–pipette method for the silt fraction (2–63 µm) of dispersed and undispersed silty soil samples (see Tables I and II for statistical details). o, Dispersed silty soil samples (Beuselinck *et al.*, 1998): Undispersed silty soil samples; _____, RMA relationship for undispersed samples; - - - - -, RMA relationship for dispersed samples

DISCUSSION

Table II gives the RMA relationships for dispersed silty soil samples (Beuselinck *et al.*, 1998). The slopes of the RMA lines of the dispersed and the undispersed silty soil samples are only significantly different for the 4–8 µm fraction. For all other fractions, the slopes and the intercepts of the RMA lines do not differ significantly. Correlation coefficients for the undispersed silty soil samples are in the same range as those for the dispersed soil samples. Thus, although there is a difference in pretreatment of the samples, RMA relationships for the dispersed and undispersed samples are very similar, as can be seen in Figures 1, 2 and 3. This is rather surprising, as it might be expected that the micro-aggregates have a lower density than individual grains, as is the case for macro-aggregates (Foster *et al.*, 1985). It also indicates that vibration of the soil sample during wet sieving and shaking of the sediment column before pipette analysis, as well as the continuous pumping of the suspension during the measurement with the Coulter LS-100, have a minor effect on micro-aggregation.

Size distributions of aggregated sediment are often used to determine the degree of aggregation both in space and time, of the sediment or the soil (e.g. Muggler *et al.*, 1997; Walling, 1990). In such studies, different techniques have been applied to measure the size distribution of the aggregated sediment. The effect of the technique used to determine aggregate size distributions is usually not taken into account. Nevertheless, the latter may be rather important. Slattery and Burt (1995) note that the degree to which the clay fraction is transported in dispersed form is important for our understanding of the fate of

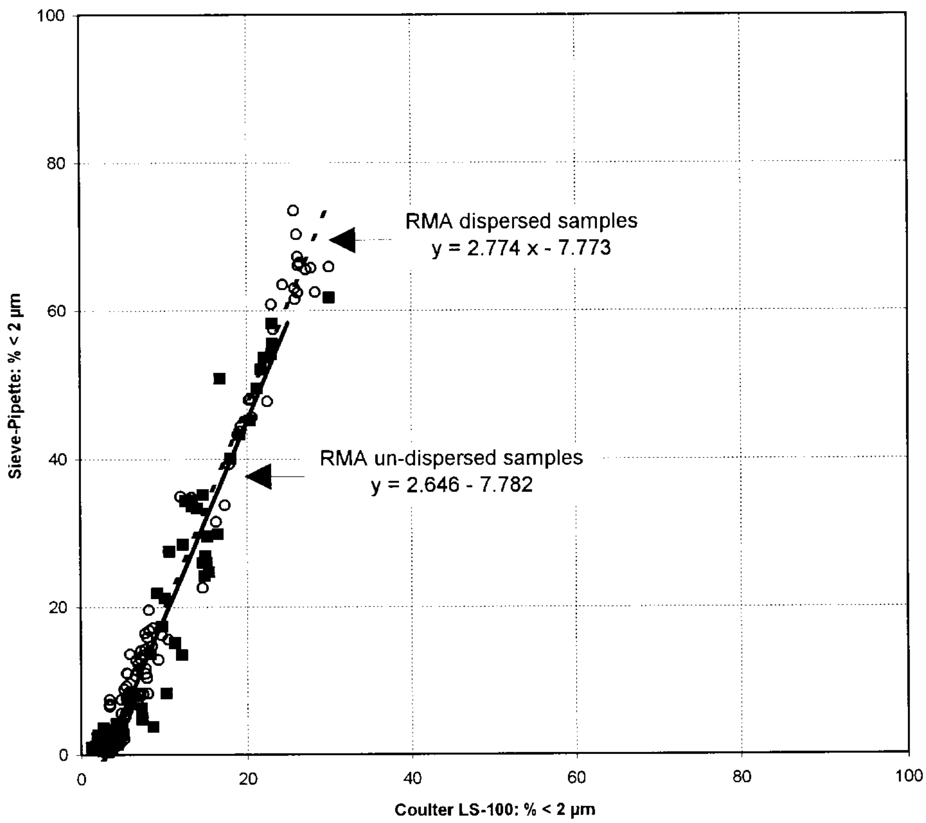


Figure 3. Comparison of the reduced major axis relationships between the Coulter LS-100 and the sieve-pipette method for the clay fraction (<2 µm) of dispersed and undispersed silty soil samples (see Tables I and II for statistical details). o, Dispersed silty soil samples (Beuselinck *et al.*, 1998); ■, undispersed silty soil samples; _____, RMA relationship for undispersed samples; - - - - -, RMA relationship for dispersed samples

sediment-associated nutrients and pollutants. The (relative) degree of dispersion of the clay fraction may be estimated as the ratio of the percentage of clay in the undispersed sample to the percentage of clay in the dispersed sample. Figure 4 shows that the estimated degree of dispersion will be strongly dependent on the method used for size analysis: for the sample shown, a much higher degree of dispersion will be obtained if the Coulter LS-100 data are used (54 per cent vs 22 per cent for the sample shown). This is due to the strong underestimation of both the dispersed and the undispersed clay fraction by the Coulter LS-100.

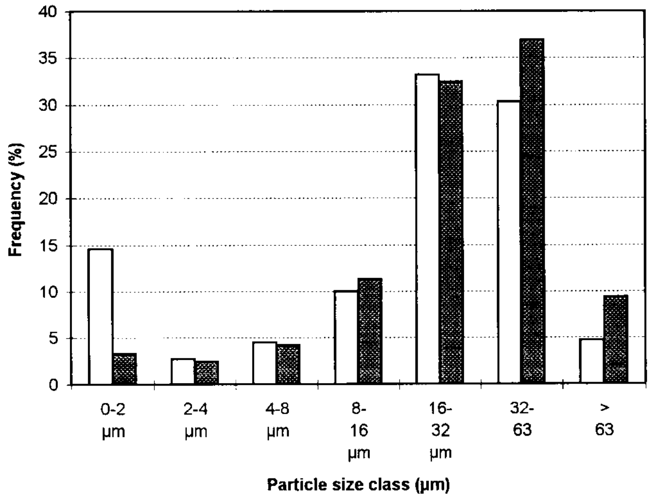
The proposed calibration relationships may be used to compare the degree of dispersion measured by both methods as a function of clay content. The overestimation of the degree of dispersion by the Coulter LS-100 is dependent on the clay fraction of the sample and on the degree of dispersion as seen by the sieve–pipette method, as shown in Figure 5. Overestimation is strongest for samples with a low clay content and with a high degree of aggregation.

Other measures of aggregation or dispersion will also be affected by the method used. A minimum aggregation index (MAI) can be calculated by summing all positive percentages after subtraction of the dispersed from the undispersed grain size fractions:

$$\mathrm{MAI} = \sum_{i=1}^{n} (D_i - U_i) \text{ for } (D_i - U_i) > 0 \tag{1}$$

where D_i = dispersed grain size percentage, U_i = undispersed grain size percentage and i = grain size class. Figure 6 shows that the MAI is clearly dependent on the measurement technique used.

(a)



(b)

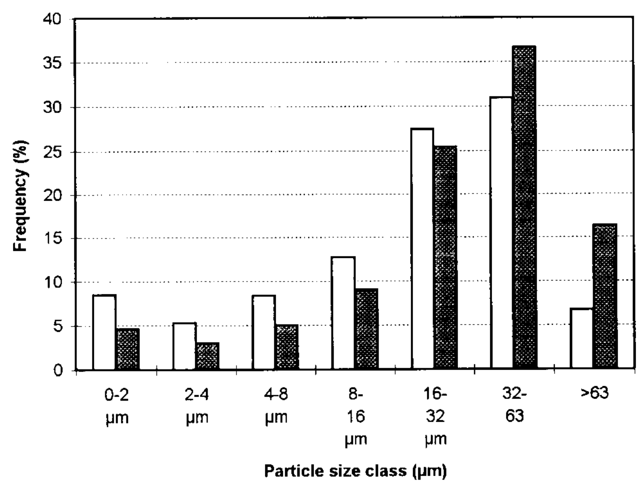


Figure 4. Undispersed (■) and dispersed (□) grain size distribution of a silty soil sample as measured by (a) the sieve–pipette method and (b) by the Coulter LS-100

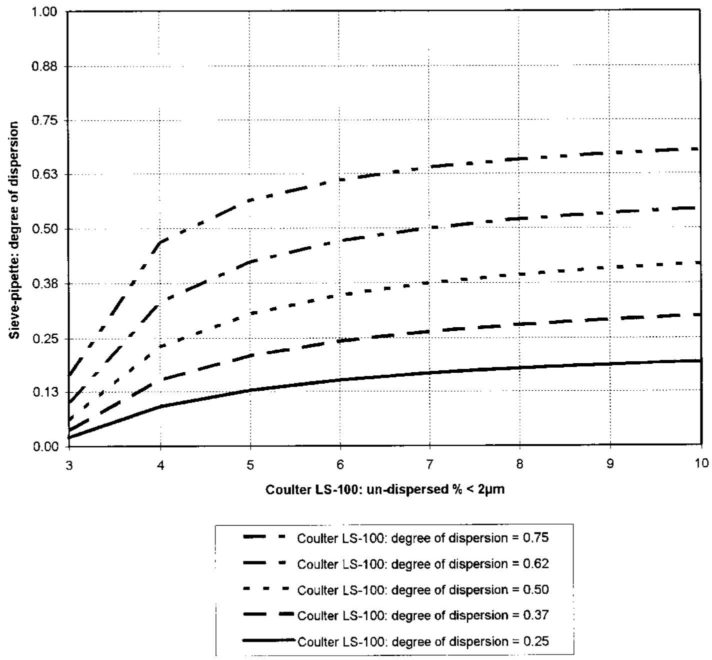


Figure 5. Nomograph indicating the degree of dispersion (percentage $<2\,\mu m$ undispersed sample / percentage $<2\,\mu m$ dispersed sample) measured with the sieve–pipette method using undispersed clay content (percentage $<2\,\mu m$) and degree of dispersion based on Coulter LS-100 data as input

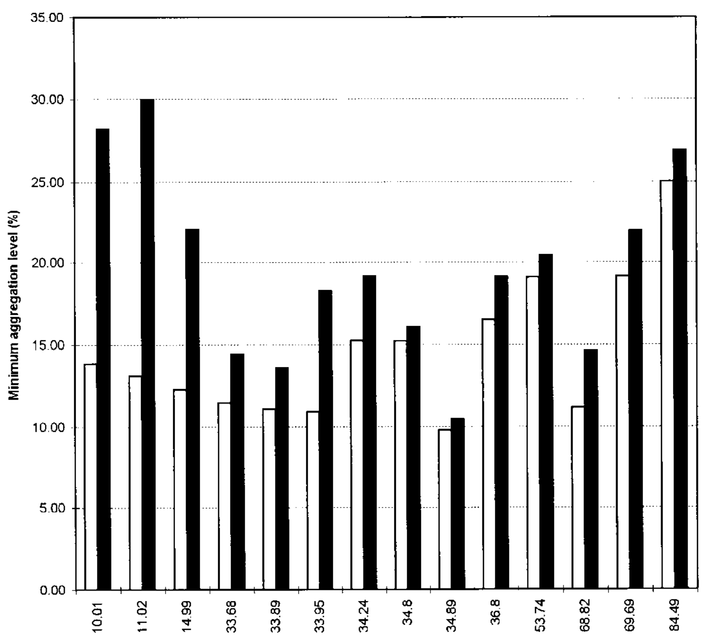


Figure 6. Minimum aggregation level measured by the Coulter LS-100 (■) and by the sieve–pipette method (□) for 15 samples sorted by increasing median of the undispersed sample (Coulter LS-100)

In summary, there exists no 'best' method for grain size analysis (Syvitski *et al.*, 1991). Our data show that both the sieve–pipette method and laser diffractometry may be used to determine the size distribution of (micro-)aggregated soil or sediment samples. Use of laser diffractometry has important practical advantages but it should be realised that the interpretation of the results of ultimate and effective particle size analyses in terms of the degree of aggregation of the sample is strongly dependent on the method used for particle size analysis. If results obtained with different methods have to be compared, a calibration is required.

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